Part 1 Point-to-point responses to the reviewers

We appreciate the reviewer's valuable comments and point-to-point responses are given below. The original comments are in black, while our responses are in blue.

RC1

1. Emission inventory: do the authors estimate CO emissions as well or they were obtained from other studies?

Response: In this study, we didn't develop emission inventory for CO. The CO emission we used in this study is from EDGAR v3 in global simulations, which is overwritten by INTEX-B (http://mic.greenresource.cn/intex-b2006) in the nested domain of East Aisa.

We add this information in 3.2 Model description in the manuscript.

2. The model evaluation. The authors used NMB as an indicator, which could potentially be affected by the compensation of overestimation and underestimation of CTM. I suggest them provide NME for Figs 3 and 4.

Response: As suggested, we calculated the NME for Fig. 3 and 4 in the original manuscript. For Fig. 3, the NME of simulated $PM_{2.5}$ concentrations in NEC, NC and YRD regions are estimated to be 38%, 45% and 36%, which is the same as the value of NMB, as the model underestimated the $PM_{2.5}$ concentration throughout the year. In MYR, SCB and PRD regions, the NME are estimated to be 18%, 21%, 22%, which are higher than the estimated NMB, especially in SCB. Overall, the model can reproduce the monthly variation of ambient $PM_{2.5}$ concentration in these key regions.

For Fig. 4, the NME of sulfate, nitrate, ammonium, BC and OC are estimated to be 58%, 41%, 28%, 44% and 50%. The NME of nitrate and ammonia show large difference with NMB. The difference mainly arise from the discrepancies between simulation and observation in NC and MYR.

In addition, we add the comparison of simulated $PM_{2.5}$ speciation with observation data averaged during 2012-2013 (X. Zhang et al., 2015), which is shown in Figure R1. The information of each site is described in detail in Zhang et al. (2012). The sulfate is underestimated by 40.5%, which mainly occurs in the two cities of Zhengzhou and Xi'an, two orange spots in central and north China, as these two sites are located in urban area. Nitrate and ammonia are overestimated by around 20%, which is a common issue in most CTMs. OC is underestimated by

28.9% due to the incomplete mechanism of SOA simulation. The NME is calculated between 30% and 41%. Generally the model can reproduced the special distribution of $PM_{2.5}$ speciation.

We also add the text and figures in 3.3 Model evaluation in the manuscript as suggested.

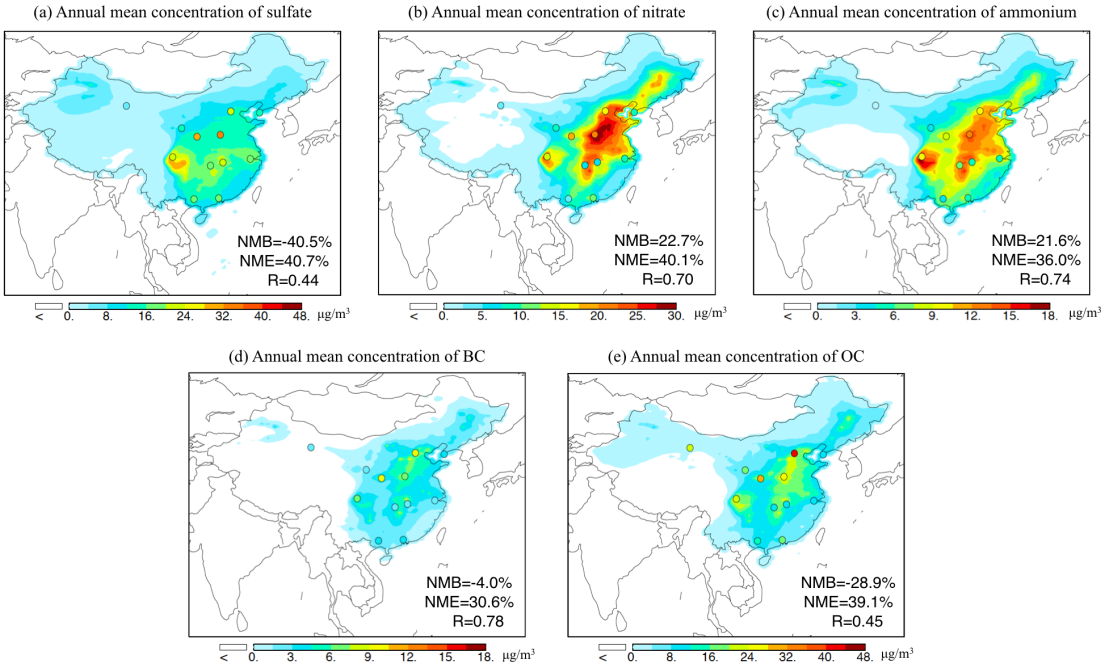

Figure R1 Comparison of simulated $PM_{2.5}$ composition with observation

3. More discussions should be given in uncertainty analysis. For example, the authors discussed the uncertainty of emission estimations based on Monte-Carlo simulation. However, it was not sufficient for readers to know the impacts of emission inventory estimation on the source apportionment results. More comparisons between various inventory studies are encouraged here to indicate the potential uncertainty of source apportionment from emission side. Moreover, there are some studies using the methods other than Brute-force to reduce the impacts of non-linear response of $PM_{2.5}$ concentrations to precursor emissions, and they should be included in the part.

Response: We looked into recent studies on major pollutant emissions in China and summarized them in Table R1. Emissions from Liu et al. (2016), Xia et al. (2016) and Wu et al. (2016) are also estimated using bottom-up method, while those from Zhao et al. (2014) are projected emissions for 2015 based on the year of 2010. We can

see that the results of this study fall into the range of previous studies except for MEP (2014) which is at low end. One major reason for low NOx emission from MEP (2014a) is that it does not include the emissions from non-road vehicles.

Table R1 Comparisons with other studies on recent air pollutant emissions in China (kt)

	SO_2	NO_X	PM_{10}	$PM_{2.5}$	VOCs
This study	23150	25638	16521	12155	23366
MEP, 2014	20439	22273	-	-	-
Liu et al., 2016	-	28300	-	-	-
Xia et al., 2016	23014-26884	28002-28817	-	-	-
Wu et al., 2016 (2012)*	-	-	-	-	29850
Zhao et al., 2014 (2015)*	26792	27511	15599	11419	-

* The year of emission are marked in brackets when it is different from the year of emission (2013) in our study.

Regarding to the non-linearity of atmospheric chemistry, there are some studies using different methods to study the source apportionment of ambient $PM_{2.5}$. As this study only focuses on coal-burning emissions in each sector, the results are not directly comparable to most similar studies except for results for power sector, as coal combustion dominates the emissions in power plant. Zhao et al. (2015) used the extended response surface modeling (ERSM) technique to access the non-linear response of fine particles to precursor emissions in each sector in PRD region, reporting that local $PM_{2.5}$ concentration decreased less than 3% (7.2% in our study) in January and around 12% in august (13.8% in our study) when 90% of emissions in power plants are reduced. Our results include the trans-boundary contributions as we shut off emissions across the country in the sensitivity simulation, which is one of the reasons causing the discrepancies. L. Zhang et al. (2015) took the advantage of the adjoint capability of GEOS-Chem, reporting that power plants contributed 6% to $PM_{2.5}$ concentration in Beijing, which is consistent with our study (6.9%).

We also add the above text in the manuscript as suggested.

4. In general the language is clear, however there are some grammar errors which need to be carefully revised before publication.

Response: We proofread the manuscript and revised grammar errors in the text carefully, as suggested.

RC2

1. The description of the simulation is too concise for the reader to understand from the information provided if the contributions to $PM_{2.5}$ from sources outside the nested domain are accounted for or not. If these contributions are accounted for, a paragraph should discuss the importance of these contributions and a Figure should show the relative importance of the sources within the domain and contrast them with outside

Response: In the nested simulation for East Asia, the contribution from outside the nested domain are accounted for. In order to quantify this contribution, we conducted another sensitivity simulation with all sources outside the domain shut off. The standard and sensitivity simulation results are shown in Fig.R2 (a) and (b), and the difference between them is analyzed as the contribution from outside the domain, which is shown in Fig.2R (c). The maximum contribution from outside is up to 13.8 μg/m^3, which mainly occurs in the west and northwest boundaries. The average contributions is 1.57 μg/m^3 in the simulation domain of East Asia. Within the boundary of China, the largest contribution occurs in the Northeast, which is 7.35 μg/m^3. The average contribution from outside the nested domain is only 0.3 μg/m^3 within China

We also add the above text and figures in 4.1 in the manuscript as suggested.

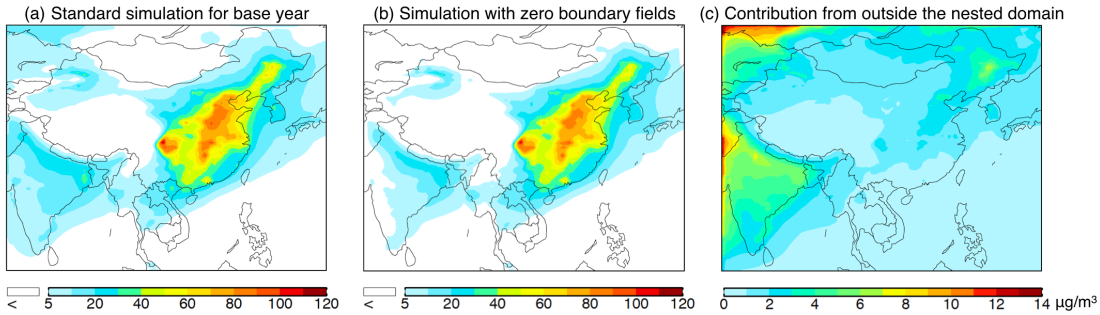

Figure R2 Contributions from outside the nested domain

2. The aerosol composition used page 6 has been gathered for measurements taken from 2006-2007 in cities across China. Your study centers on the year 2013. How did you connect this composition for 2006-2007 to the year 2013?

Response: We didn't adjust the observation during 2006-2007 to connect to our simulation, but took into account the differences of emissions in 2006, 2007 and 2013, when we interpret the evaluation results. To better resolve this issue, we add the evaluation using the observation data from X. Zhang et al. (2015), which is shown in Figure R3. The observed concentration is the average from 2012 to 2013. The information of each site is described in detail in Zhang et al. (2012). The underestimate of sulfate mainly occurs in the two cities of Zhengzhou and Xi'an, two orange spots in central and north China, as these two sites are located in urban area. Nitrate and ammonia are overestimated by around 20%, which is a common issue in most CTMs. OC is underestimated by 28.9% due to the incomplete mechanism of SOA simulation. The NME is calculated between 30% and 41%. Generally the model can reproduced the special distribution of $PM_{2.5}$ speciation.

We add the text and figures in 3.3 Model evaluation in the manuscript.

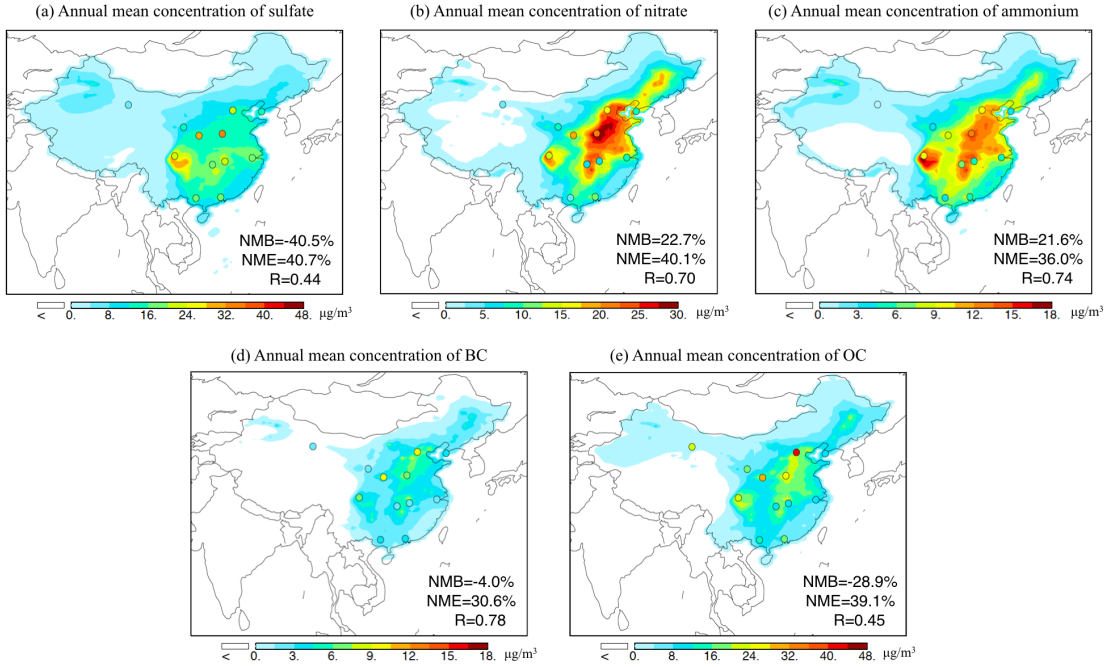

Figure R3 Comparison of simulated $PM_{2.5}$ composition with observation

3. Concerning Figure 2, you present the maps of surface $PM_{2.5}$ for four seasons and simply give the normalized mean bias and the correlation coefficient. I would like to see with Figure 2 the correlation plots so that the reader can have a better view of how the predicted $PM_{2.5}$ concentrations agree/disagree with the measured ones.

Response: We made the correlation plot for each season, as shown in Fig. R4. The $PM_{2.5}$ concentration is more spread out in coordinates in winter as it varies substantially across China, which has a larger correlation coefficient of 0.71. In other seasons, the correlation coefficients are around 0.6.

We also add the above text and figures in 3.3 Model evaluation in the manuscript as suggested.

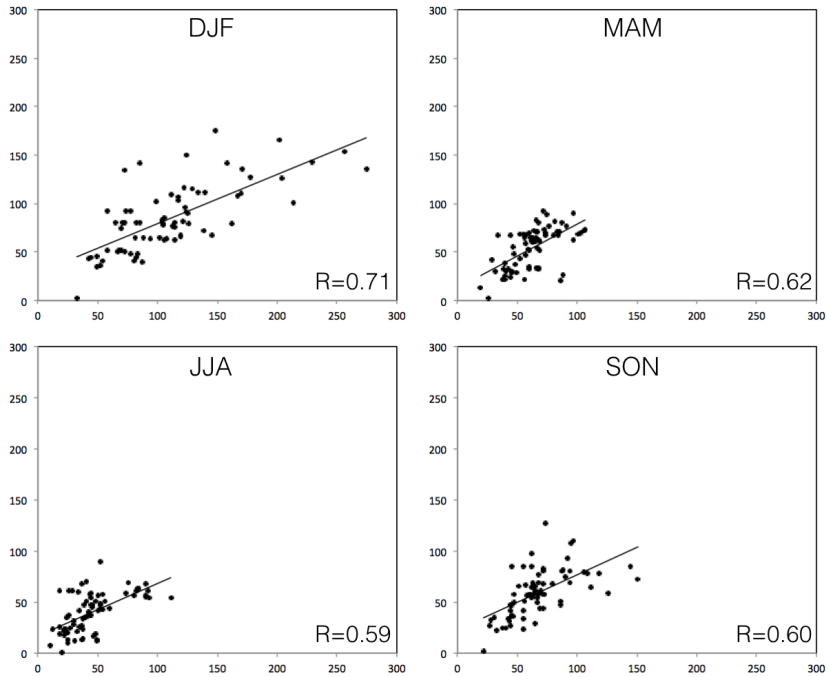

Figure R4 Correlation maps for four seasons

4. Finally the syntax for paragraphs 4.1 through 4.4 should be improved before the manuscript is considered for publication in ACP

Response: We re-plot the figures from 4.1 to 4.4 and improved the syntax in the manuscript as suggested.

Part 2 List of relevant changes in manuscript

1. We add the information of CO emission in 3.2 Model description as per comment #1 in RC 1.
2. We add the NME of data in Figure 3 as per comment #2 in RC 1.
3. We add the comparisons of emissions in this study with others in Table 7 and the relevant illustration in 4.4 Uncertainty analysis as per comment #3 in RC 1.
4. We add the comparisons of our source apportionment results with studies using other methods in 4.4 Uncertainty analysis as per comment #3 in RC 1.
5. We corrected several grammar errors in as per comment #4 in RC 1.
6. We add the contribution from outside the nested domain as Figure 6 and the relevant illustration in 4.4 as per comment #1 in RC2.
7. We add the correlation maps for $PM_{2.5}$ concentration in each season as Figure 3 and the relevant illustration in 3.3 Model evaluation as per comment #2 in RC 2.
8. We substitute observation data during 2012-2013 for the data during 2006-2007 in model evaluation of $PM_{2.5}$ speciation in Figure 5 as per comment #3 in RC 2. The related description is also modified.
9. We re-plot Figure 5, Figure 7 and Figure 8, and improved the syntax as per comment #4 in RC 2.
10. We add the following references according to the above modifications.

Liu F, Zhang Q, Zheng B, et al. Recent reduction in NOx emissions over China: synthesis of satellite observations and emission inventories [J]. Environmental Research Letters, 2016, 11(11): 114002.

Zhao B, Wang S X, Xing J, et al. Assessing the nonlinear response of fine particles to precursor emissions: development and application of an extended response surface modelling technique v1. 0[J]. Geoscientific Model Development, 2015, 8(1): 115-128.

Wu R, Bo Y, Li J, et al. Method to establish the emission inventory of anthropogenic volatile organic compounds in China and its application in the period 2008–2012[J]. Atmospheric Environment, 2016, 127: 244-254.

Xia Y, Zhao Y, Nielsen C P. Benefits of China's efforts in gaseous pollutant control indicated by the bottom-up emissions and satellite observations 2000–2014[J]. Atmospheric Environment, 2016, 136: 43-53.

Zhang X Y, Wang J Z, Wang Y Q, et al. Changes in chemical components of aerosol particles in different haze regions in China from 2006 to 2013 and contribution of meteorological factors[J]. Atmospheric Chemistry and Physics, 2015, 15(22): 12935-12952.

Zhao Y, Zhang J, Nielsen C P. The effects of energy paths and emission controls and standards on future trends in China's emissions of primary air pollutants [J]. Atmospheric Chemistry and Physics, 2014, 14(17): 8849-8868.

Part 3 Marked-up manuscript

Impacts of Coal Burning on Ambient $PM_{2.5}$ Pollution in China

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Abstract. High concentration of fine particles ($PM_{2.5}$), the primary concern about air quality in China, is believed to closely relate to China's large consumption of coal. In order to quantitatively identify the contributions of coal combustion in different sectors to ambient $PM_{2.5}$, we developed an emission inventory for the year 2013 using up-to-date information on energy consumption and emission controls, and conducted standard and sensitivity simulations using the chemical transport model GEOS-Chem. According to the simulation, coal combustion contributes 22 μg m^{-3} (40%) to the total $PM_{2.5}$ concentration at national level (averaged in 74 major cities), and up to 37 μg m^{-3} (50%) in Sichuan Basin. Among major coal-burning sectors, industrial coal burning is the dominant contributor with a national average contribution of 10 μg m^{-3} (17%), followed by coal combustion in power plants and domestic sector. The national average contribution due to coal combustion is estimated to be 18 μg m^{-3} (46%) in summer and 28 μg m^{-3} (35%) in winter. While the contribution of domestic coal burning shows an obvious reduction from winter to summer, contributions of coal combustion in power plants and industrial sector remain at relatively constant levels through out the year.

1 Introduction

$PM_{2.5}$ (particulate matter with aerodynamic diameter less than or equal to 2.5 μm), was considered as the leading air pollutant in most key regions and cities in China, especially in the Beijing-Tianjin-Hebei (BTH) region and the Yangtze River Delta (YRD), according to the air quality status reports released by China's Ministry of Environmental Protection

(MEP, 2014a; MEP, 2015). The annual mean $PM_{2.5}$ concentration in BTH region was 102 μg m^{-3} in 2013 and 93 μg m^{-3} in 2014, while that in YRD was 67 μg m^{-3} in 2013 and 60 μg m^{-3} in 2014 (MEP, 2014a; MEP, 2015), far beyond ~~even~~ the World Health Organization (WHO) interim target-1 (35 μg m^{-3}) for annual mean $PM_{2.5}$ concentration and also the secondary class standard in the China's new National Ambient Air Quality Standard (NAAQS, GB 3095-2012).

The high ambient $PM_{2.5}$ concentration is believed to closely relate to China's large primary energy consumption, especially coal consumption. According to BP statistical review of world energy (BP, 2015), China has become the largest energy consumer since 2009, and coal occupied 2/3 of the total primary energy consumption~~of which 2/3 is coal consumption~~. In the year 2010, coal was responsible for ~~more than~~ 81% of the SO_2 emissions, 61% of the NO_X emissions, 40% of the primary PM_{10} emissions, and 34% of the primary $PM_{2.5}$ emissions in China (S. Wang et al., 2014b). As the most abundant and a relatively cheap energy resource, coal is expected to be a dominant energy supply in China in the foreseeable future.

A number of studies have used atmospheric models to study the source contributions of energy use to ambient air pollution in China. Early studies (Wang et al., 2005; Hao et al., 2007) mainly focused on gaseous pollutants, including SO_2, NO_X, CO and O_3. Later on, more studies (Bi et al., 2007; Cheng et al., 2007; Chen et al., 2007; Hao et al., 2007; Wang et al., 2008; Wu et al., 2009) place emphasis on particulate matter, but mainly on PM_{10}. Recently, due to the frequent haze episodes characterized by extremely high $PM_{2.5}$ concentration in China, researchers pay more and more attention to $PM_{2.5}$. Among these studies, most of them took the advantage of 3-D chemical transport models like Community Multi-scale Air Quality Model (CMAQ). H. Zhang et al. (2012) studied source contributions to sulfate and nitrate in $PM_{2.5}$ using the CMAQ model and reported that while power sector is the largest contributor to inorganic components, industry and traffic sector are also important sources. Some recent studies agreed that industrial and domestic sources were the most significant contributors to ambient $PM_{2.5}$ in most areas in China. L. Wang et al. (2014) studied a severe $PM_{2.5}$ pollution episode in Jan. 2013 in North China ~~for Hebei province~~ using the CMAQ model and concluded that industrial and domestic sources respectively contributed 28% and 27% to local $PM_{2.5}$ concentration in Hebei province. D. Wang et al (2014) conducted simulations with the same model and studied the same pollution episode but for a different city of Xi'an in northwestern China, also reporting that industrial and domestic activities are the two largest sources that account~~s~~ for 58% and 16% respectively. L. Zhang et al. (2015) used the GEOS-Chem model and indicated that the residential and industrial sources in North China were respectively responsible for 49.8% and 26.5% of the $PM_{2.5}$ concentration in Beijing. While most of the studies focused on developed metropolises or heavy pollution episodes, very few studies used atmospheric chemical transport models to study source contributions and its seasonal variation for the whole country throughout a year. In addition, while most researchers studied the total energy consumption in each sector or regarded coal combustion in all sectors as a whole, none of them distinguished coal burning in one sector from another. However, the utilization of coal and the end-of-pipe emission control policies are quite different in ~~each~~ different sectors, which leads to different energy efficiency and thus different emissions. Therefore, contributions from coal burning in specific sectors should be identified respectively, which is important for policy making.

In this study, we updated a previously developed emission inventory to the year 2013 using up-to-date information and conducted sensitivity simulations with the chemical transport model GEOS-Chem. In order to obtain a comprehensive understanding of the current contribution from coal combustion to $PM_{2.5}$ concentrations in China, we quantitatively identified source contributions from coal burning in each sector and its seasonal variation. Section 2 discusses the development of emission inventory for the year 2013; section 3 describes the method of simulation, GEOS-Chem model and its evaluation; section 4 discusses the model results; the last section summarizes the conclusions.

2 Emission inventory

Our previous studies have developed the emission inventory of sulfur dioxide (SO_2), nitrogen oxide (NO_X), PM_{10}, $PM_{2.5}$, black carbon (BC), organic carbon (OC), non-methane volatile organic compounds (NMVOC), and ammonia (NH_3) for China for the year 2010 using a *technology-based emission factor method* (S. Wang et al., 2014b; Zhao et al., 2013a; Zhao et al., 2013b; Zhao et al., 2013c). The emissions from each sector in each province were calculated from the activity data (energy consumption, industrial products, solvent use, etc.), technology-based emission factors, and penetrations of control technologies. In this study, we updated the 2010 emission inventory to year 2013 by incorporating the most recent information. The activity data and technology distribution for each sector were updated to 2013 according to the Chinese Statistics (NBS, 2014a; NBS, 2014b; NBS, 2014c) and a wide variety of technology reports (Fu et al., 2015; S. Wang et al., 2014b; CEC, 2011; ERI, 2010; ERI, 2009; THUBERC, 2009). The emission factors used in this inventory were described in Zhao et al. (2013b). The penetrations of removal technologies were updated to 2013 according to governmental bulletins and the evolution of emission standards (MEP, 2014b).

There are some significant updates for NH_3 emissions in this inventory. For agricultural fertilizer application, the emissions of NH_3 in the previous study were based on pre-defined emission factors that lacked temporal or spatial details ~~in previous studies~~. In this inventory, we use an agricultural fertilizer modeling system that couples the regional air quality model CMAQ and an agro-ecosystem model (the Environmental Policy Integrated Climate model, EPIC) to improve the accuracy of spatial and temporal distribution (Fu et al., 2015). For livestock, the activity data were calculated by the amount of livestock slaughter per year in previous studies. However, the survival periods for each livestock are different and not only one year, thus the amount of slaughter cannot stand for the amount of livestock accurately. In this study, we use the amount of livestock stocks to calculate NH_3 emissions and improve the accuracy of the results.

In 2013, the anthropogenic emissions of SO_2, NOx, PM_{10}, $PM_{2.5}$, BC, OC, NMVOC and NH_3 in China were estimated to be 23.2 Mt, 25.6 Mt, 16.5 Mt, 12.2 Mt, 1.96 Mt, 3.42 Mt, 23.3 Mt, and 9.62 Mt, respectively. Table 1 shows emissions by sector and emissions originating from coal combustion, which indicates that in sectors of power plants and domestic fossil fuel combustion, the share of coal-burning emissions are almost over 90%. Coal dominates the emissions in industrial sector as well. In the year of 2013, coal is responsible for 79% of the SO_2 emissions, 54% of the NO_X emissions, 40% of the primary PM_{10} emissions, and 35% of the primary $PM_{2.5}$ emissions, 40% of the BC emissions and 17% of the OC emissions.

3 Model and simulation

3.1 Simulation method

In this study, we conducted one standard simulation and 4 sensitivity simulations for ground level $PM_{2.5}$ using the nested grid capability of GEOS-Chem for East Asia. The simulation scenarios are summarized in Table 2. In the standard simulation, we use the emissions for the year 2013 that are discussed in Section 2. To select the year of meteorology, we conducted standard simulation using the same emissions and different meteorology from the year 2010 to 2012, as the meteorological fields are not available for the whole year of 2013. We chose the year 2012 as our meteorology year, with which the simulation results best represented the mean $PM_{2.5}$ concentration from 2010 to 2012.

In sensitivity scenarios, we respectively removed emissions from coal combustion in different sectors. In sensitivity scenario 1, we removed emissions from coal burning from all energy sectors (scenario for total coal burning, TC). In sensitivity scenarios 2 to 4, we respectively shut down emissions from coal burning in power plants, industries and domestic sectors (TCP, TCI and TCD). All the meteorology used in the sensitivity simulation was the same as the standard simulation. Three months before each simulation year were used as spin-up. The differences between standard and sensitivity simulations are used to represent the contributions from coal combustion in each sector.

3.2 Model description

GEOS-Chem is a global chemical transport model that has been widely applied to study $PM_{2.5}$ over China (e.g. Brauer et al., 2012, 2015; Jiang et al., 2015; Kharol et al., 2013; van Donkelaar et al., 2010, 2015; Y. Wang et al., 2013, 2014; Xu et al. 2015; L. Zhang et al. 2015; Q. Zhang et al., 2015). The model is driven by assimilated meteorological data from the United States National Aeronautics and Space Administration (NASA) Goddard Earth Observing System (GEOS), including winds, temperature, clouds, precipitation, and other surface properties. GEOS-Chem (version 9-01-03) includes detailed HO_X-NO_X-VOC-ozone-BrO_X tropospheric chemistry originally described by Bey et al. (2001a) with addition of BrO_X chemistry by Parrella et al. (2012). Aerosol simulation is fully coupled with gas-phase chemistry, including sulfate (SO_4^{2-}), Nitrate (NO_3^-), and ammonium (NH_4^+)(Park et al., 2004; Pye et al., 2009), OC and BC (Park et al., 2003), sea salt (Alexander et al., 2005), and mineral dust (Fairlie et al., 2007). The areasol thermodynamic equilibriums use the ISORROPIA II model (Fountoukis and Nenes, 2007) to calculate the partitioning of nitric acid and ammonia between gas and aerosol phases. The formation of secondary organic aerosol (SOA) includes the oxidation of isoprene (Henze and Seinfeld, 2006), monoterpenes, aromatics (Henze et al., 2008) and other reactive VOCs (Liao et al., 2007). In addition, we corrected errors in the model representation of too shallow nighttime mixing depth following Walker et al. (2012) and introduced the production mechanism of sulfate on aerosol surface described in Wang et al. (2013). Aerosols interact with gas-phase chemistry in GEOS-Chem through the effect of aerosol extinction on photolysis rates (Martin et al., 2003) and heterogeneous chemistry (Jacob, 2000).

In this study, we conducted simulations for ground level $PM_{2.5}$ using the nested grid capability of GEOS-Chem for East Asia, which was originally described by Wang et al. (2004) and Chen et al. (2009). The nested domain for East Asia covers area

spanning from 70°E to 150°E, and from 11°S to 55°N, with a horizontal resolution of 0.5 latitudes by 0.667 longitudes. The boundary fields are provided by the global GEOS-Chem simulation with a resolution of 4 latitudes by 5 longitudes and are updated every 3 hours. We assume that the organic mass/organic carbon ratio is 1.8 and relative humidity is 50% for $PM_{2.5}$ in China.

The global simulations use emissions from the Global Emission Inventory Activity (GEIA) inventory (Benkovitz et al., 1996), which is respectively overwritten by the NEI05, EMEP and INTEX-B inventory (Zhang et al., 2009) over the US, Europe, and East Asia. The CO emission we used in this study is from EDGAR v3, which is also overwritten by INTEX-B in the nested domain of East Asia. In the nested-grid simulation for East Asia, we use the emissions for the year 2013 as discussed in Section 2 over China, with emissions over the rest of East Asia taken from the INTEX-B emission inventory. In addition, the simulation also includes open fire emissions from GFED3 inventory (Giglio et al., 2010; van der Werf et al., 2010; Mu et al. 2011), lightning NO_X emissions calculated with algorithm of Prince and Rind (1992), volcanic SO_2 emissions from AEROCOM data base (http://www-lscedods.cea.fr/aerocom/AEROCOM_HC/) implemented by Fisher et al. (2011).

3.3 Model evaluation

GEOS-Chem model is driven by assimilated meteorological data from the NASA GEOS. Y. Wang et al. (2014) has evaluated the important meteorological factors that are relevant to particle formation in the model, including temperature, relative humidity (RH), wind speed and direction, using observation data from National Meteorological Information Center (NMIC) of China. It reported good spatial and temporal correlations with observed temperature, RH and wind direction. The correlation of wind speed, however, was poorer as the model tends to overestimate in low speed conditions.

In this study, we conducted model evaluation using the surface $PM_{2.5}$ observation network of China National Environmental Monitoring Center (CNEMC, http://106.37.208.233:20035). This monitoring program was initiated in January 2013, covering 74 major cities in China. Fig. 1 compares simulated annual mean $PM_{2.5}$ concentrations with those observed in 74 major cities in China for the year 2013. As shown in Fig. 1a, the simulated ambient $PM_{2.5}$ concentration has a clear regional distribution with high values in the Sichuan Basin (SCB), North China Plain (NC), and middle Yangtze River area (MYR). The highest concentration occurs in Sichuan Basin with an average value of 73.5 μg m^{-3}. Concentrations in the above-mentioned severely polluted regions are generally above 60 μg m^{-3}. The observation data isare compared with the concentrations in ~~of~~ the grids where the city centers are located. The comparison shows that model well reproduces the spatial distribution with a normalized mean bias (NMB) of -16.3%. The correlation coefficient for annual mean concentration is 0.68. The slight underestimate mainly appears in the heavily polluted area in NC region where observations are largely influenced by local emissions but current simulation cannot capture it with a relatively coarse resolution (H. Zhang et al., 2012). Fig. 2 shows comparisons between simulated and observed seasonal mean concentrations. $PM_{2.5}$ concentration has an obvious seasonal variation with the highest value in winter and the lowest in summer, which is correctly reproduced by the model. The largest bias occurred in winter with the value of -23.3%. The inconsistency of meteorology ~~field~~ also partly accounts for the underestimate, as the meteorology condition was more unfavorable in Jan. 2013. Y. Wang

et al. (2014) conducted simulations for Jan. in 2012 and 2013 using same emissions, and found the ground $PM_{2.5}$ concentration was ~~are~~ 27% higher in Jan. 2013 than that in 2012. Model performs better in other three seasons, with biases between -13.3% and -10.8%. Correlation maps for each season are shown in Fig.3. The $PM_{2.5}$ concentration in winter is more spread out in coordinates as it varies substantially across China, which has a larger correlation coefficient of 0.71. In other seasons, the correlation coefficients are around 0.6.~~The seasonal correlation coefficients varied between 0.59 and 0.71.~~

We also evaluated the monthly variation using averaged monthly mean concentrations in cities in each key region, as analyses and discussions mainly focused on these six areas. The six key regions are shown with frames in Fig. 1a, which includes Northeast China (NEC, 123°E-128°E, 41°N-47°N), North China (NC, 113°E-119°E, 33°N-40°N), Yangtze River Delta (YRD, 119°E-122°E, 29.5°N-32.5°N), Middle Yangtze River (MYR, 111°E-115°E, 27°N-32.5°N), Sichuan Basin (SCB, 103°E-107°E, 28°N-32°N) and Pearl River Delta (PRD, 112°E-114°E, 22°N-24°N). Cities in each region share the similar weather condition, terrain and pollution levels. As shown in Fig. 4~~3~~, the model generally well reproduces the monthly variation. The NMB ranges from -45% to 1%, and the correlation coefficient varies between 0.7 and 0.94. The model performance is better in MYR, SCB and PRD than that in NC, NEC and YRD. The large discrepancy is mainly due to the failure to capture the extremely high concentration in wintertime. The normalized mean errors (NME) of simulated $PM_{2.5}$ concentrations in NEC, NC and YRD regions are estimated to be 38%, 45% and 36%, which is the same as the values of NMB, as the model underestimated the $PM_{2.5}$ concentration throughout the year. In MYR, SCB and PRD regions, the NME are estimated to be 18%, 21%, 22%, which are higher than the estimated NMB, especially in SCB. Overall, the model can reproduce the monthly variation of ambient $PM_{2.5}$ concentration in these key regions.

The $PM_{2.5}$ composition shows a great diversity across China. Sulfate-nitrate-ammonium (SNA), BC, Organic Matter (OM), and crustal material respectively constituted 7.1% to 57%, 1.3% to 12.8%, 17.7% to 53% and 7.1% to 43% in $PM_{2.5}$ mass in China, and the fractions of SNA in $PM_{2.5}$ (40% - 57%) is much higher in East China (Yang et al., 2011). OM and mineral dust also play significant roles in $PM_{2.5}$ concentration. $PM_{2.5}$ speciation in China simulated by GEOS-Chem has been evaluated in some previous studies. Wang et al. (2013) reported annual biases of -10%, +31%, and +35% for sulfate, nitrate and ammonia respectively, compared with observations at 22 sites in East Asia. Fu et al. (2012) indicated that annual mean BC and OC concentrations in rural and background sites were underestimated by 56% and 75%. $PM_{2.5}$ speciation is also evaluated in this study using the observed concentration of aerosol compositions averaged from 2012 to 2013 in 12 cities across China (X. Zhang et al., 2015), as shown in Fig.5. The information of each site is described in detail in Zhang et al. (2012). The underestimate of sulfate mainly occurs in the two cities of Zhengzhou and Xi'an, two orange spots in central and north China, as these two sites are located in urban area. Nitrate and ammonia are overestimated by around 20%, which is a common issue in most CTMs. OC is underestimated by 28.9% due to the incomplete mechanism of SOA simulation. The NME is calculated between 30% and 41%. The correlation coefficients range between 0.44 and 0.78. ~~$PM_{2.5}$ speciation is also evaluated in this study using the observed concentration of aerosol compositions from 2006 to 2007 in 16 cities across China (X. Zhang et al., 2012), which is shown in Fig.4. The model underestimates the annual mean concentration of sulfate, ammonium, BC and OC by 58%, 13%, 34% and 49% respectively, and overestimates nitrate concentration by 2%. The~~

~~correlation coefficients range between 0.71 and 0.84. However, the SO_2 emissions were estimated to be 27826 kt in 2006 and 26455 kt in 2007, and it decreased to 23129 kt in 2013 (S. Wang et al., 2014b). In contrast, the NO_x emissions increased to 25623 kt in 2013 from 20791 kt in 2006 and 22287 kt in 2007 (Zhao et al., 2013c). Considering the evident change of SO_2 and NO_x emissions in China from 2006 to 2013, the underestimate for sulfate should be less than 58% and the overestimate for nitrate is higher that 2%.~~

4 Source contributions to ambient $PM_{2.5}$ concentration

4.1 Annual mean source contributions

Fig. 6~~5~~ shows the spatial distribution of annual mean source contributions from coal burning. As shown in Fig. 5~~6~~a, the contribution from total coal burning has a similar spatial distribution with the annual mean $PM_{2.5}$ concentration, which indicates the large influence of coal burning on air quality. Table 3 also shows a higher percentage contribution in areas with higher $PM_{2.5}$ concentrations such as NC, MYR and SCB regions. The national average contribution from total coal burning, which is an average of concentrations in 74 major cities, is up to 22.5 μg m^{-3}, ~~which~~ accounting~~s~~ for almost 40% of the total $PM_{2.5}$ concentration. In the six key regions, coal burning contributes 34.5% to 50.2% of the total ambient $PM_{2.5}$ concentration. The largest contribution occurs in SCB, which reaches 36.9 μg m^{-3} on average, due to the dense population, large emissions and unfavorable terrain that tends to trap the emissions and secondary pollutants in this area. The highest contribution is up to 56.9 μg m^{-3}, occurring in the southwest city of Chengdu. Following SCB, coal-burning contributions in MYR and NC are also above the national average, with average values of 30.8 μg m^{-3} (45.1%) and 26 μg m^{-3} (40.5%), respectively. Among the six key regions, coal combustion in PRD shows the smallest contribution of 12.6 μg m^{-3}, yet still accounting for 35% of the local $PM_{2.5}$ concentration. In addition to the key regions, coal burning contributes to around 25 μg m^{-3} (more than 50%) of the local $PM_{2.5}$ in cities like Baotou and Hohhot in Inner Mongolia, an autonomous region near the middle north border, as it is one of the largest production areas of coal and a large amount of raw coal is burnt for energy supply. In the northwest city of Urumqi, coal burning is also a large contributor for it accounts for around 40% of the local $PM_{2.5}$ concentration as there are no other large anthropogenic sources of air pollutants there.

Among all the subsectors in coal combustion, industrial coal burning is the most significant contributor, followed by coal burning in power plants and domestic sector, which is shown in Fig. 5~~6~~b - d and Table 3. The contribution from industrial coal burning is up to 9.6 μg m^{-3} (17%) on national average (74 major city average), while those from coal burning in power plants and domestic sector are 5.6 μg m^{-3} (9.8%) and 2.2 μg m^{-3} (4%), respectively. The contribution from each sector differs in different regions. Contributions from coal burning in power plants and industry have similar spatial distributions with the annual mean $PM_{2.5}$ concentration. As shown in Fig. 5~~6~~b, coal burning in power plants has the largest contribution in NC with the highest value of 13.1 μg m^{-3} (15%) and an average of 7.7 μg m^{-3} (12%), due to the large number of power plants in this area. The smallest contribution occurs in PRD with the value of only 2.7 μg m^{-3} (7.5%). In most key areas in China, coal burning in power sector contributes to around 10% of the local $PM_{2.5}$ concentration, which is a relatively minor source

compared with industry due to higher energy efficiency and more stringent emission control policies in power sectors. Industrial coal burning, as shown in Fig. ~~5~~6c, has the largest contribution in SCB, with an average value of 19 μg m^{-3} (25.9%). The largest contribution occurs in the city of Chengdu, which is up to 35.8 μg m^{-3}, accounting for around 1/3 of the local $PM_{2.5}$. NC and MYR are also significantly influenced by industrial coal burning with the contributions of 10.8 μg m^{-3} (16.8%) and 14 μg m^{-3} (20.5%), respectively. In other areas including NEC, YRD and PRD, the average contributions of coal burning in industrial sector are generally less than 10 μg m^{-3}, accounting for around 15% of the local $PM_{2.5}$ concentration. As shown in Fig. ~~5~~6d, domestic coal burning has little contribution to ambient $PM_{2.5}$ in most areas in the six key regions. However, in some individual regions in Guizhou province in Southwest and Inner Mongolia in North China, domestic coal burning contributes more than 10 μg m^{-3}, which accounts for more than 15% in Guizhou and 25% in Inner Mongolia where people tend to burn more raw coal for heating. Besides, the high sulfur content of coal in Guizhou province also accounts for the large contribution.

In the nested simulation of East Asia, the contribution from outside the nested domain are also accounted for. In order to quantify the back ground concentration, we conducted another sensitivity simulation with all sources outside the domain shut off. The standard and sensitivity simulation results are shown in Fig.7 (a) and (b), and the difference between them is analysed as the contribution from outside the domain, which is shown in Fig.7 (c). The maximum contribution from outside is up to 13.8 μg/m^3, which mainly occurs in the west and northwest boundaries. The average contributions is 1.57 μg/m^3 in the simulation domain of East Asia. Within the boundary of China, the largest contribution occurs in the Northeast, which is 7.35 μg/m^3. The average contribution from outside the nested domain is only 0.3 μg/m^3 within China.

4.2 Seasonal variation of coal contributions

Fig. ~~8~~6 shows the simulated seasonal mean $PM_{2.5}$ concentration (Fig. ~~8~~6a and b) and source contributions from coal burning in winter (averaged from December to February) and in summer (averaged from June to August) (Fig. ~~8~~6c to j), which is also summarized in Table 4 and 5. As shown in Fig. ~~8~~6a and b, the ambient $PM_{2.5}$ concentration has obviously different distributions in winter and in summer. $PM_{2.5}$ in winter has a similar distribution with the annual mean, but with much higher values. The highest value still occurs in SCB with an average of 118.8 μg m^{-3} due to the large emission, unfavorable terrain and weather condition in winter. Following SCB, the average concentrations in MYR and NC regions are above 100 μg m^{-3} and 90 μg m^{-3}, respectively. There are also several populated cities in NEC, where $PM_{2.5}$ are generally above 75 μg m^{-3} and up to 150 μg m^{-3}. $PM_{2.5}$ in summer has an obviously different distribution from winter with much lower concentrations and more even distribution through out the country due to the stronger vertical mix, more wet deposition and lower emissions. The largest concentration occurs in NC region with 46.9 μg m^{-3} on average, followed by SCB with an average of 44.1 μg m^{-3}. In addition to the above two regions, $PM_{2.5}$ concentrations in other key regions are generally around or below 35 μg m^{-3} on average.

In winter, coal burning contributes to 28.2 μg m^{-3} (35.4%) of~~to~~ total $PM_{2.5}$ concentration on the national level. Similar with the annual mean, coal-burning contribution in winter peaks in SCB with an average of 50.3 μg m^{-3} (42.3%) and reaches the

lowest in PRD with 16.1 μg m^{-3} (29%). Among the coal-burning sectors, the contributions from power plants and industry also have similar spatial pattern with the annual mean distribution. Coal burning in industry, followed by that in power plants, is the largest contributor in both seasons. Domestic coal burning is a significant contributor in winter, due to the large amount of emissions from heating supply. The high $PM_{2.5}$ concentration from domestic sector mainly occurs in some areas in Guizhou Province in southwest and Inner Mongolia in north, where a large amount of raw coal is burnt for heating. The largest contribution reaches as much as 37.6 μg m^{-3} in Inner Mongolia, which accounts for almost 40% of the local $PM_{2.5}$ concentration.

In summer, the national average contribution from coal burning is estimated to be 17.8 μg m^{-3} (46.2%), which is less than 2/3 of the contribution in winter, due to the favorable meteorological condition including stronger convection and more frequent wet deposition. Regional contribution ranges from 8.2 μg m^{-3} in PRD to 26.3 μg m^{-3} in SCB, which is approximately half of the contributions in winter. The seasonal variation of contributions in inland areas (NEC, MYR, SCB) is more significant than those in coastal areas (NC, YRD, PRD). In coal-burning sectors, the absolute contributions from power plants and industry doesn't show very noticeable reductions in summer compared with those in winter, as emissions from these two sectors are in a relatively constant status throughout the year and the nitrate reduction due to the high temperature in summer is counteracted by the enhancement of the sulfate formation (H. Zhang et al., 2012). In contrast, domestic sector contributes 1 μg m^{-3} (2.5%) on the national level in summer, which is 3 to 8 times less than that in winter.

4.3 Comparisons with other studies

The Natural Resources Defense Council (NRDC) launched the China Coal Consumption Cap Project in Oct. 2013 and released the report of *contribution of coal use to air pollution in China* as part of the study results in October 2014 (NRDC, 2014). This study used the CAMx model with MEIC inventory and meteorology from WRF to simulate coal contributions to ambient $PM_{2.5}$ in January, February, April and October in the year 2012 in 333 main cities in China. In order to compare with the NRDC study, we extracted the simulated contribution in the 333 main cities during the same periods from our study results. Fig. 9~~7~~ represents the comparison in each province and shows that our study underestimates the coal contribution by 22% compared to that in the NRDC study. The discrepancy is mainly generated from the different amounts of emissions that are originated from coal in the two studies. According to the report, the NRDC study included both emissions directly from coal burning and emissions from industries closely related to coal burning. For example, air pollutants from industries like coke, steel, cement and non-ferrous metal are generated from two ways: directly from coal combustion and from the technological process. As coal is used as fuel in these industries and is not likely to be substituted for in the near future, the NRDC study includes both the two parts as emissions from *coal use*. In our study, we include only the first part of the emissions as the contribution from coal, which is actually generated from *coal burning*. According to the report by NRDC, coal combustion is responsible for 79% of the SO_2 emissions, 57% of the NO_X emissions and 44% of the primary PM emissions, and the coal-related sources are responsible for 15%, 13% and 23% of the SO_2, NO_X and PM emissions. Despite

of the difference definition of coal contribution to air pollutant emissions, the NRDC and our study both predicted high contribution to $PM_{2.5}$ concentration from coal, especially in the Municipality of Chongqing and Sichuan province in SCB.

4.4 Uncertainty analysis

The uncertainties of the contribution estimates in this study may arise from the uncertainties of the emission inventory, model simulation and non-linearity of the atmospheric chemistry. A Monte Carlo uncertainty analysis was performed on the emission inventory, as described in Zhao et al. (2013c) and S. Wang et al. (2014b). Table 6 shows the uncertainty analysis of the emissions in China. Among all the coal-consuming sectors analyzed in this study, domestic sector is subject to the highest uncertainty, which may lead to more uncertainty in the $PM_{2.5}$ simulation and contribution estimates. Other studies on major pollutant emissions in China are summarized in Table 7. Emissions from Liu et al. (2016), Xia et al. (2016) and Wu et al. (2016) are also estimated using bottom-up method, while those from Zhao et al. (2014) are projected emissions for 2015 based on the year of 2010. The results of this study fall into the range of previous studies except for MEP (2014) which is at low end. One major reason for low NOx emission from MEP (2014a) is that it does not include the emissions from non-road vehicles.

Another important cause of uncertainty is the model simulation of the $PM_{2.5}$ composition. The coal contribution to sulfate is larger than that to nitrate, as the share of coal-burning emissions of SO_2 is 79% in this study, 25% higher than that of NO_X emissions. Therefore, the actual coal-burning contribution to $PM_{2.5}$ is very likely to be larger than the estimates in this study, due to the underestimation of sulfate concentration and overestimation of nitrate concentration by the model.

In addition, due to the non-linear response of $PM_{2.5}$ concentration to precursor emissions, contributions from coal burning in each sector add up to less than the contribution from the total coal burning, which means the probable underestimation of the contribution in subsectors. The impact of non-linearity of the atmospheric chemistry on $PM_{2.5}$ concentrations and its composition has been discussed in detail in previous studies (Zhao et al., 2013b; S. Wang et al., 2014a). There are some studies using different methods to study the source apportionment of ambient $PM_{2.5}$. As this study only focuses on coal-burning emissions in each sector, the results are not directly comparable to most similar studies except for results for power sector, as coal combustion dominates the emissions in power plant. Zhao et al. (2015) used the extended response surface modeling (ERSM) technique to access the non-linear response of fine particles to precursor emissions in each sector in PRD region, reporting that local $PM_{2.5}$ concentration decreased less than 3% (7.2% in our study) in January and around 12% in august (13.8% in our study) when 90% of emissions in power plants are reduced. Our results include the trans-boundary contributions as we shut off emissions across the country in the sensitivity simulation, which is one of the reasons causing the discrepancies. L. Zhang et al. (2015) took the advantage of the adjoint capability of GEOS-Chem, reporting that power plants contributed 6% to $PM_{2.5}$ concentration in Beijing, which is consistent with our study (6.9%).

5 Conclusion

We updated China's emission inventory to the year 2013 using up-to-date information on energy statistics and emission control policies. The anthropogenic emissions of SO_2, NOx, PM_{10}, $PM_{2.5}$, BC, OC, NMVOC and NH_3 in China were estimated to be 23.2 Mt, 25.6 Mt, 16.5 Mt, 12.2 Mt, 1.96 Mt, 3.42 Mt, 23.3 Mt, and 9.62 Mt, respectively. Using the emission inventory, we conducted standard and sensitivity simulations for major coal-burning sectors to quantitatively identify the source contributions from coal burning using the chemical transport model GEOS-Chem. Results show that coal combustion contributes to 22.5 μg m^{-3} (40%) of the total $PM_{2.5}$ concentration on national average (74 major city average). The highest contribution occurs in Sichuan Basin, which reached 36.9 μg m^{-3} and accounts for more than 50% of the local $PM_{2.5}$. Among the subsectors of coal combustion, industrial coal burning is the dominant contributor, with the largest contribution of 19 μg m^{-3} (26%) in Sichuan Basin and the second largest of 14 μg m^{-3} (20%) in Middle Yangtze River area, which indicates that coal combustion in industry should be prioritized when energy policies and end-of-pipe control strategies are applied, especially in middle-west regions in China, from the perspective of the whole country. Coal combustion in power plants shows the largest contribution in North China with an average of 7.7 μg m^{-3} (12%). Domestic coal burning has the largest contribution in some regions in Guizhou province in Southwest China and Inner Mongolia in North China, where combustion of raw coal should be substantially reduced especially in winter. An obvious seasonal variation is also predicted. The absolute contributions due to coal combustion are estimated to be 28 μg m^{-3} (35%) in winter and 18 μg m^{-3} (46%) in summer on the national level. The seasonal differences are mainly due to the dramatic change of domestic emissions and more favorable meteorological conditions including stronger convection and wet deposition in summer. While contribution from domestic coal shows a significant reduction from winter to summer, the absolute contributions from coal burning in power plants and industry remain at relatively steady levels throughout the year.

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Table 2 Emissions by sector in 2013 in China (Unit: kt)

	SO_2	NOx	PM_{10}	$PM_{2.5}$	BC	OC	NMVOC	NH_3
Power plants	6275.4	6463.6	1034.2	612.1	8.1	14.9		
Coal[a]	6209.2	6091.2	1000.9	579.1	3.6	0.0		
Industrial combustion	7226.5	4399.8	1536.0	1030.1	142.8	41.2	133.5	
Coal[b]	5972.2	2969.4	1233.9	805.6	108.4	21.4	63.7	
Other industrial process	2061.1	2492.7	3173.2	1982.3	561.2	429.3	6297.4	215.0
Coal[c]	718.8	1758.9	1521.8	782.8	220.2	179.7	1188.8	
Cement	1704.0	2884.8	2985.1	1866.7	11.3	33.9		
Coal[d]	1270.8	2151.4	1224.4	843.1	8.4	25.3		
Steel	1859.8	532.6	1388.3	1024.2	37.7	48.2		
Coal[e]	1325.1	379.5	463.0	400.4	26.9	34.4		
Domestic fossil fuel combustion	2887.3	609.6	1320.9	974.4	448.1	348.5	4265.6	918.6
Coal	2692.6	554.0	1220.4	893.0	413.4	317.4	848.0	
Domestic biofuel combustion	72.4	477.9	2970.8	2878.0	503.7	1582.9		
On-road transportation	644.0	5138.2	121.2	114.8	52.4	33.5	2044.2	
Off-road transportation	329.5	2111.6	243.6	230.8	131.5	41.5	868.8	
Solvent use							8155.3	
Biomass open burning	90.2	527.1	1747.9	1441.6	57.7	576.6	1213.8	
Waste disposal							387.4	
Livestock farming								5489.8
Mineral fertilizer application								2997.9
National total emissions	**23150.2**	**25638.0**	**16521.2**	**12155.1**	**1955.1**	**3423.1**	**23366**	**9621.3**
Emissions from coal combustion	**18188.7**	**13904.4**	**6664.4**	**4304.0**	**780.9**	**578.2**	**2100.4**	

[a] Coal here refers to emissions from coal in the corresponding sector in the above row.

[b, c, d, e] In this study industrial coal combustion includes emissions from these four sector.

Table 2 Summary for simulation scenarios

		Scenarios	Description	Meteorology
Standard scenario		STD	Standard emission for the year 2013	2012
Sensitivity scenarios	1	TC	Emissions from total coal burning removed	2012
	2	TCP	Emissions from coal burning in power plants removed	2012
	3	TCI	Emissions from coal burning in industry removed	2012
	4	TCD	Emissions from domestic coal burning removed	2012

Table 3 Annual mean absolute contributions (μg m^{-3}) and percentage contributions from coal burning

	Mean $PM_{2.5}$	Total coal burning contributions		Contributions from coal burning in					
				Power plant		Industry		Domestic	
National Average*	56.7	22.5	39.6%	5.6	9.8%	9.6	17.0%	2.2	4.0%
NEC	34.5	13.2	38.3%	3.6	10.4%	5.3	15.3%	1.8	5.3%
NC	64.3	26.0	40.5%	7.7	12.0%	10.8	16.8%	1.9	2.9%
YRD	52.2	18.0	34.5%	5.1	9.8%	7.6	14.6%	0.7	1.4%
MYR	68.3	30.8	45.1%	6.9	10.1%	14.0	20.5%	2.7	3.9%
SCB	73.5	36.9	50.2%	5.6	7.6%	19.0	25.9%	4.0	5.5%
PRD	36.2	12.6	35.0%	2.7	7.5%	5.7	15.8%	0.9	2.5%

* The National average is an average of concentrations in 74 grids where major city centers are located.

Table 4 Seasonal absolute contributions (μg m^{-3}) and percentage contributions from coal burning in winter

	Mean $PM_{2.5}$	Total coal burning contributions		Contributions from coal burning in					
				Power plant		Industry		Domestic	
National Average*	79.6	28.2	35.4%	6.3	7.9%	9.4	11.8%	4.3	5.4%
NEC	53.6	20.6	38.5%	5.5	10.3%	6.8	12.7%	4.0	7.4%
NC	90.0	31.8	35.3%	9.2	10.2%	10.6	11.8%	3.1	3.4%
YRD	66.2	19.5	29.5%	4.8	7.2%	6.7	10.1%	1.2	1.7%
MYR	104.9	40.2	38.3%	9.3	8.9%	14.0	13.4%	3.8	3.6%
SCB	118.8	50.3	42.3%	7.4	6.3%	18.9	15.9%	7.3	6.2%
PRD	55.4	16.1	29.0%	2.2	4.0%	5.4	9.8%	1.8	3.2%

* The National average is an average of concentrations in 74 grids where major city centers are located.

Table 5 Seasonal absolute contributions (μg m^{-3}) and percentage contributions from coal burning in summer

	Mean $PM_{2.5}$	Total coal burning contributions		Contributions from coal burning in					
				Power plant		Industry		Domestic	
National Average*	38.4	17.8	46.2%	5.2	13.4%	9.0	23.4%	1.0	2.5%
NEC	20.3	8.9	44.1%	2.7	13.3%	4.8	23.4%	0.5	2.5%
NC	46.9	21.7	46.4%	7.3	15.5%	10.5	22.5%	1.0	2.1%
YRD	34.1	14.2	41.5%	4.7	13.8%	6.7	19.5%	0.18	0.5%
MYR	36.2	20.2	56.1%	5.1	14.2%	11.6	32.0%	1.6	4.5%
SCB	44.2	26.2	59.5%	4.7	10.7%	16.0	38.5%	1.9	4.2%
PRD	20.2	8.2	40.7%	2.2	10.8%	4.3	21.5%	0.3	1.5%

* The National average is an average of concentrations in 74 grids where major city centers are located.

Table 6: Results of the uncertainty analysis of the emissions in China.

	NO_X	SO_2	$PM_{2.5}$	NMVOC
Power plants	±34%	±30%	±31%	-
Industrial sector	±41%	±49%	±53%	±63%
Residential sector	±55%	±51%	±68%	±65%
Transportation	±66%	±48%	±52%	±57%
Solvent use	-	-	-	±78%
Other sectors[a]	±177%	±179%	±216%	±184%
Total emissions[b]	[-31%,44%]	[-29%,45%]	[-39%,49%]	[-42%,67%]

[a] Other sectors mainly refer to open biomass burning.

[b] The last line shows the average 90% confidence intervals of the total emissions.

Table 7: Comparisons with other studies on recent air pollutant emissions in China (kt)

	SO_2	NO_X	PM_{10}	$PM_{2.5}$	VOCs
This study	23150	25638	16521	12155	23366
MEP, 2014	20439	22273	-	-	-
Liu et al., 2016	-	28300	-	-	-
Xia et al., 2016	23014-26884	28002-28817	-	-	-
Wu et al., 2016 (2012)*	-	-	-	-	29850
Zhao et al., 2014 (2015)*	26792	27511	15599	11419	-

* The year of emission are marked in brackets when it is different from the year of emission (2013) in our study.

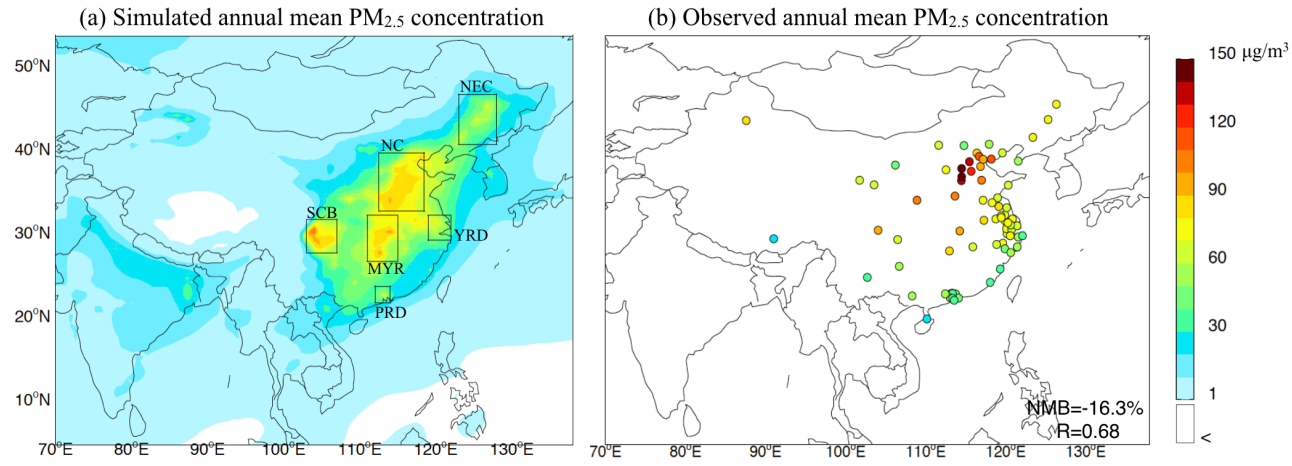


Figure 1: Simulated and observed annual mean $PM_{2.5}$ concentration in China. The six key regions include the Northeast China (NEC), North China (NC), Yangzte River Delta (YRD), Sichuan Basin (SCB), Middle Yangzte River (MYR), and Pearl River Delta (PRD).

5

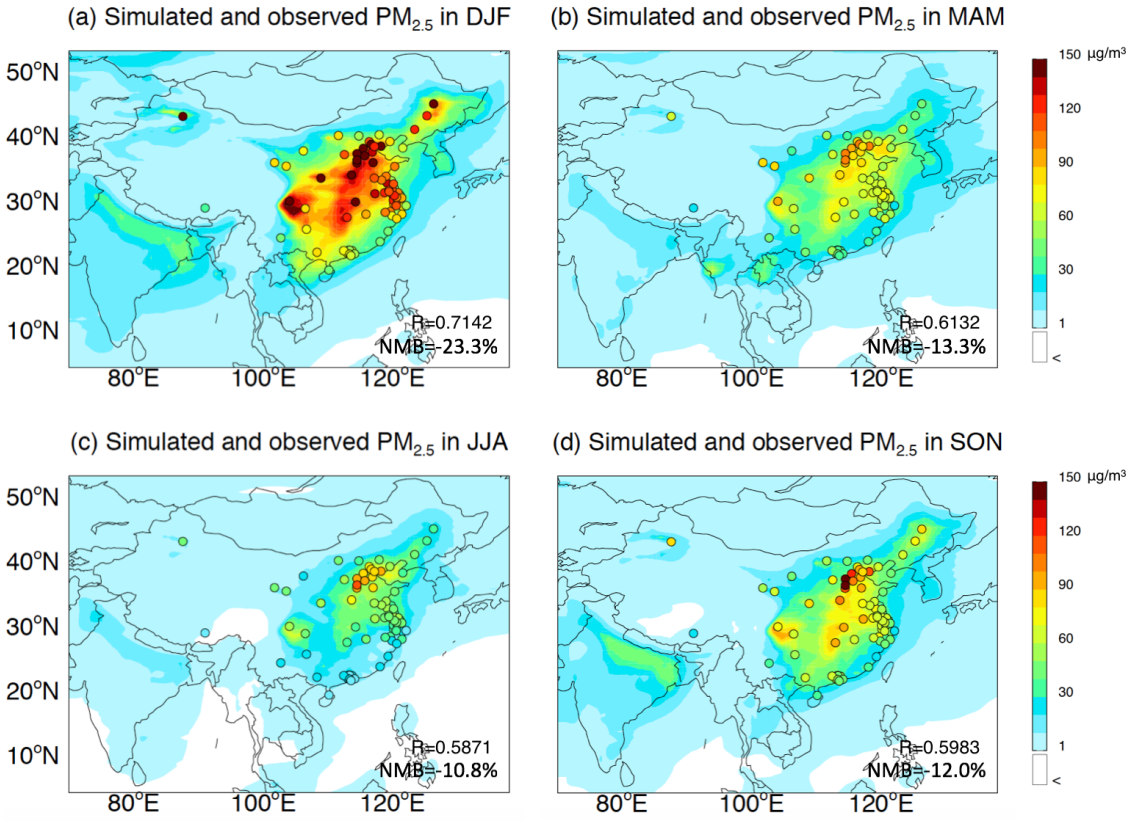


Figure 2: Simulated and observed sensonal $PM_{2.5}$ concentration in China.

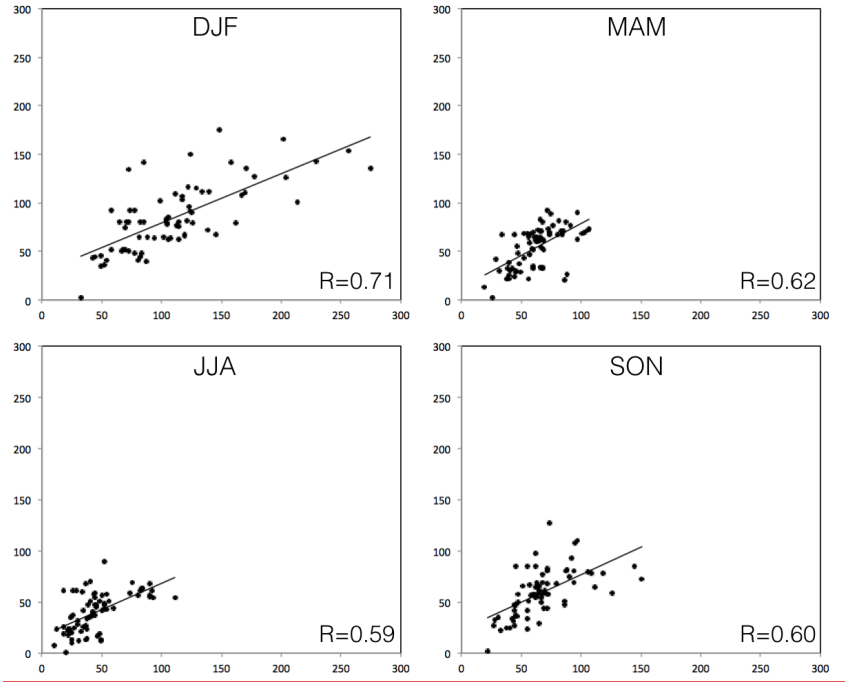


Figure 3: Correlation maps for each season.

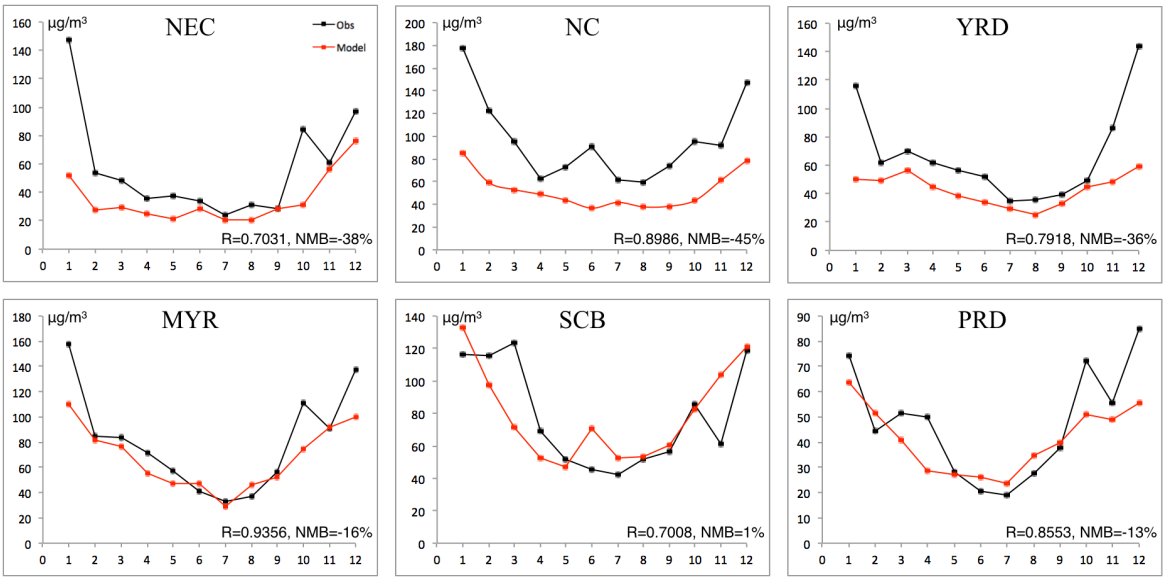
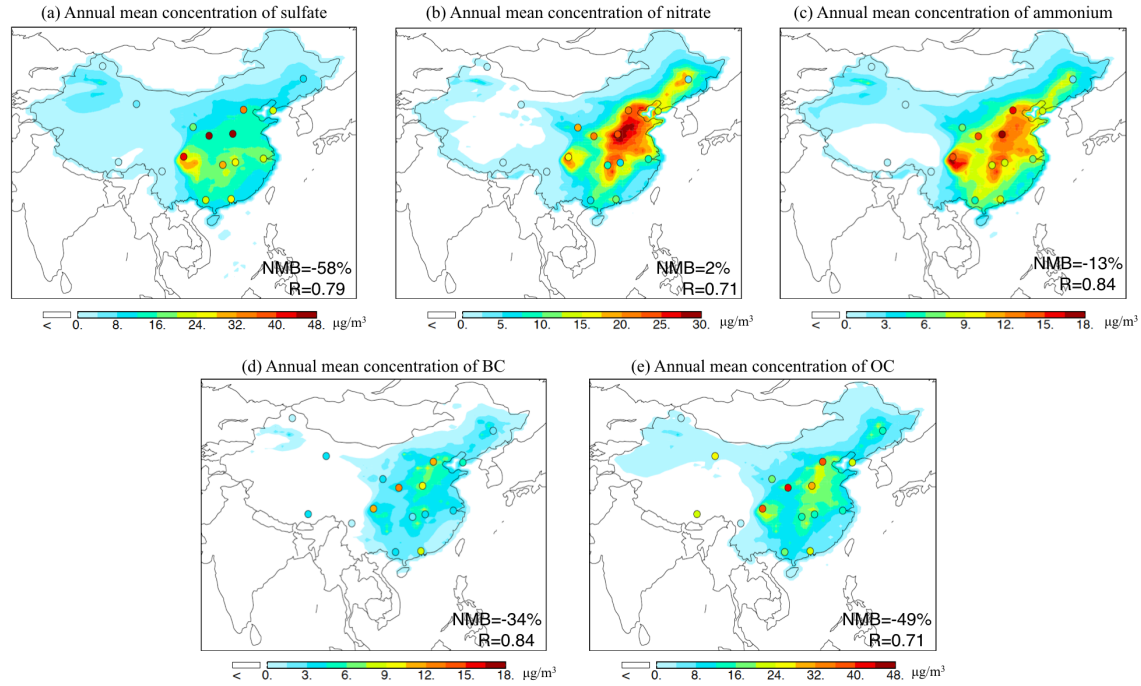


Figure ~~43~~: Monthly mean simulated and observed $PM_{2.5}$ in 6 key regions.



~~Figure 4: Simulated (2013) and observed (2006-2007) $PM_{2.5}$ speciation in China.~~

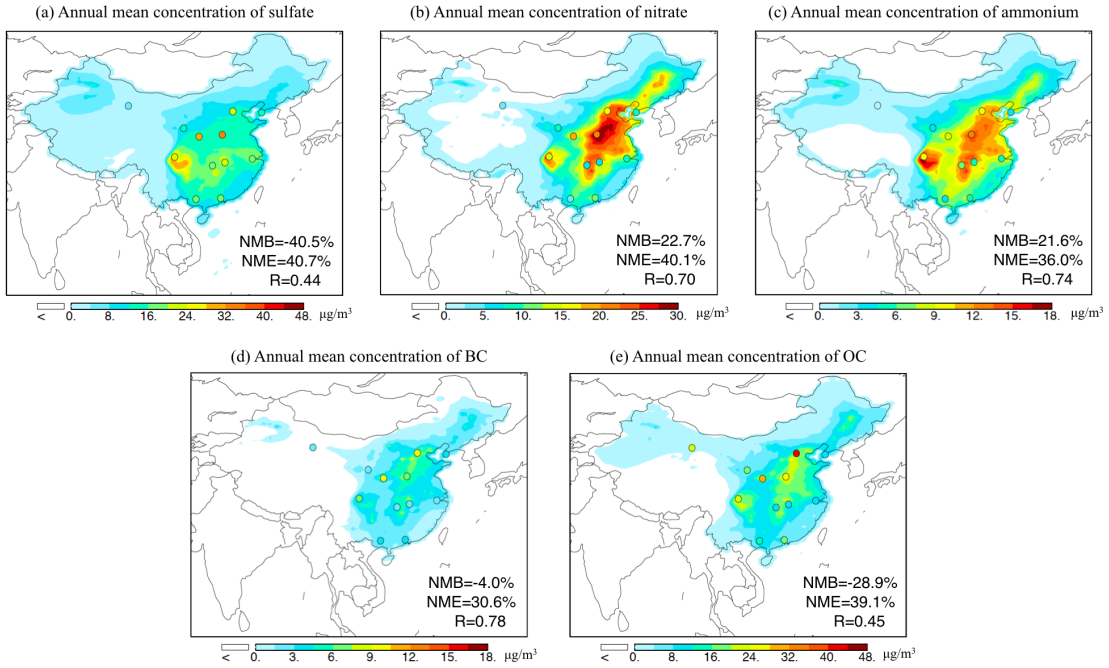
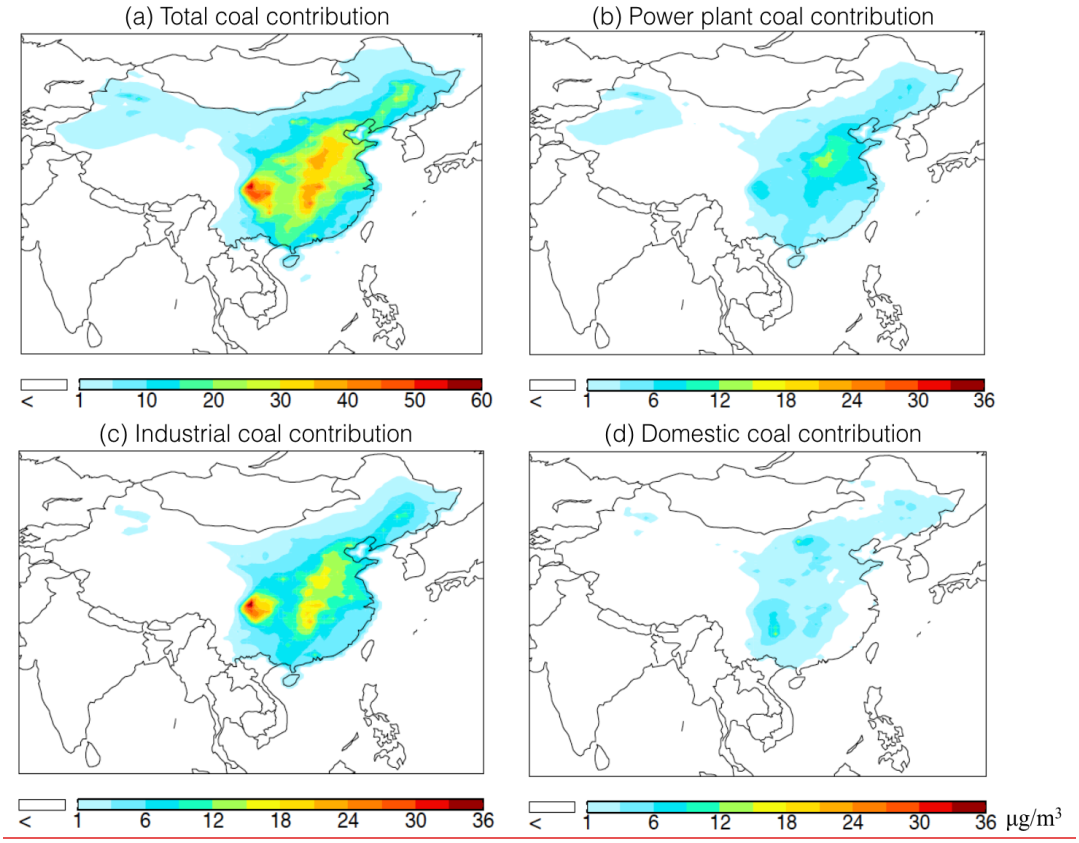


Figure 5: Simulated and observed $PM_{2.5}$ composition in China.



~~Figure 5: Annual mean contributions from coal burning.~~

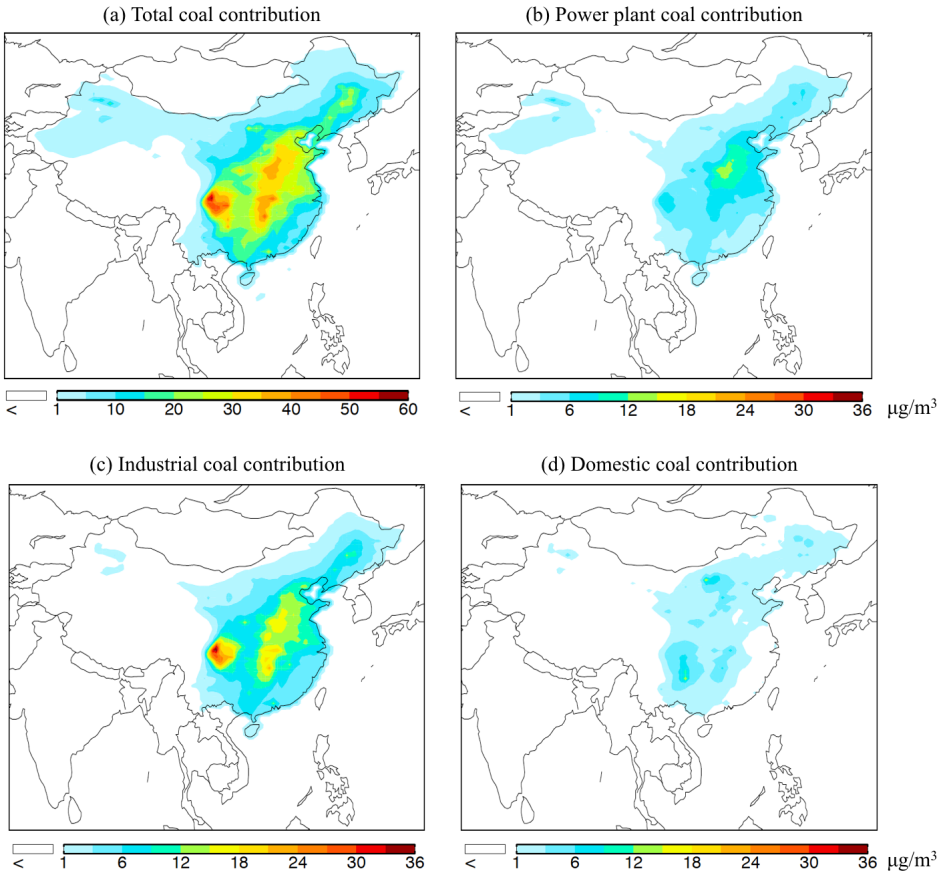


Figure 6: Annual mean contributions from coal combustion

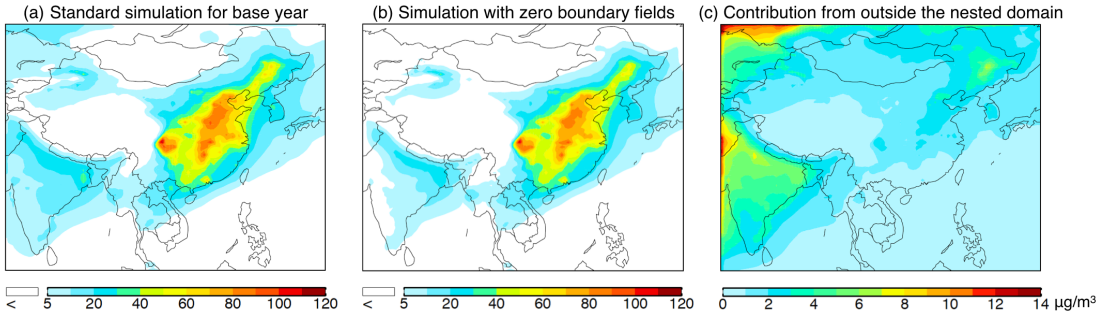
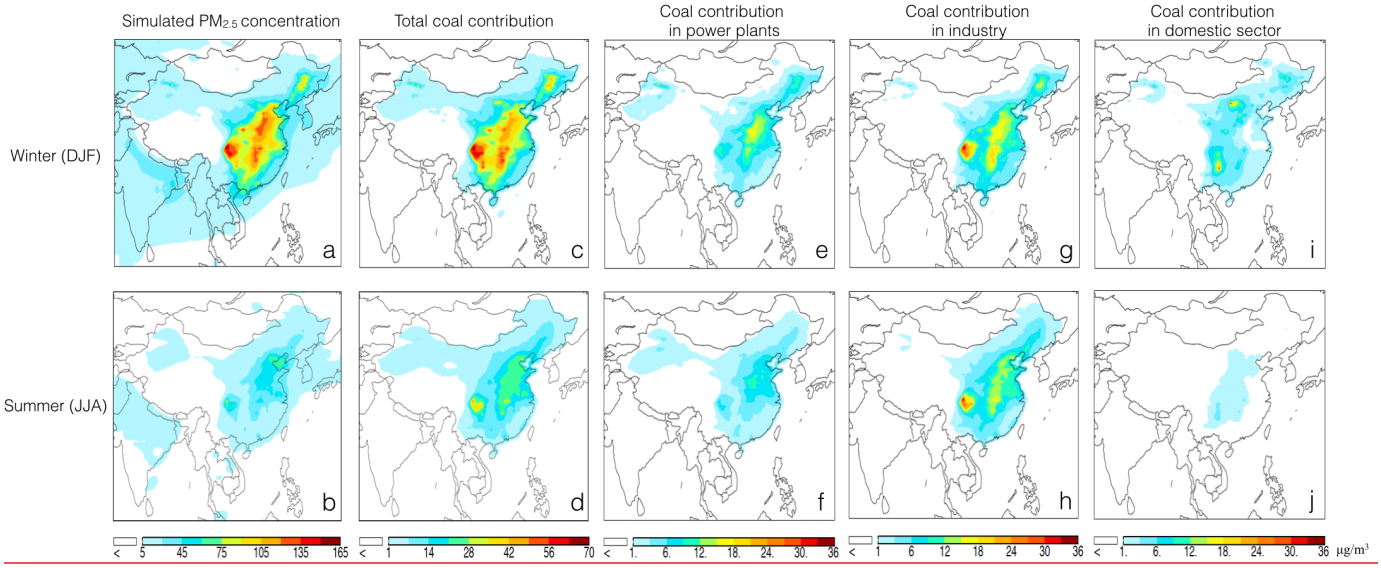


Figure 7: Annual mean contributions from outside the nested domain



~~Figure 6: Seasonal contributions from coal burning in winter and summer.~~

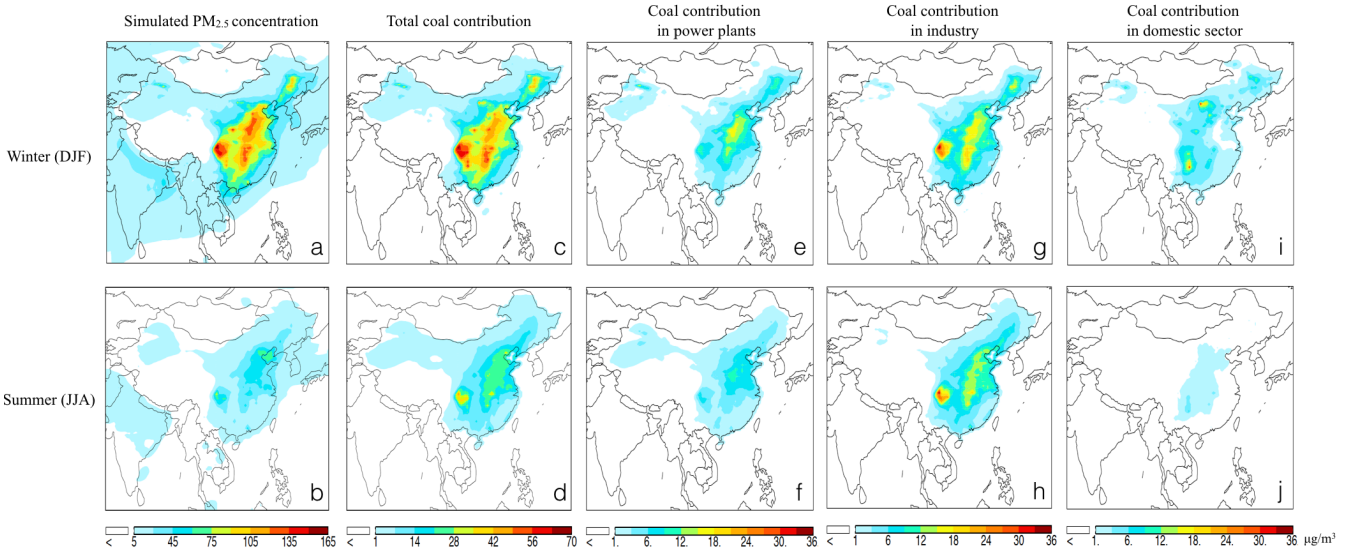
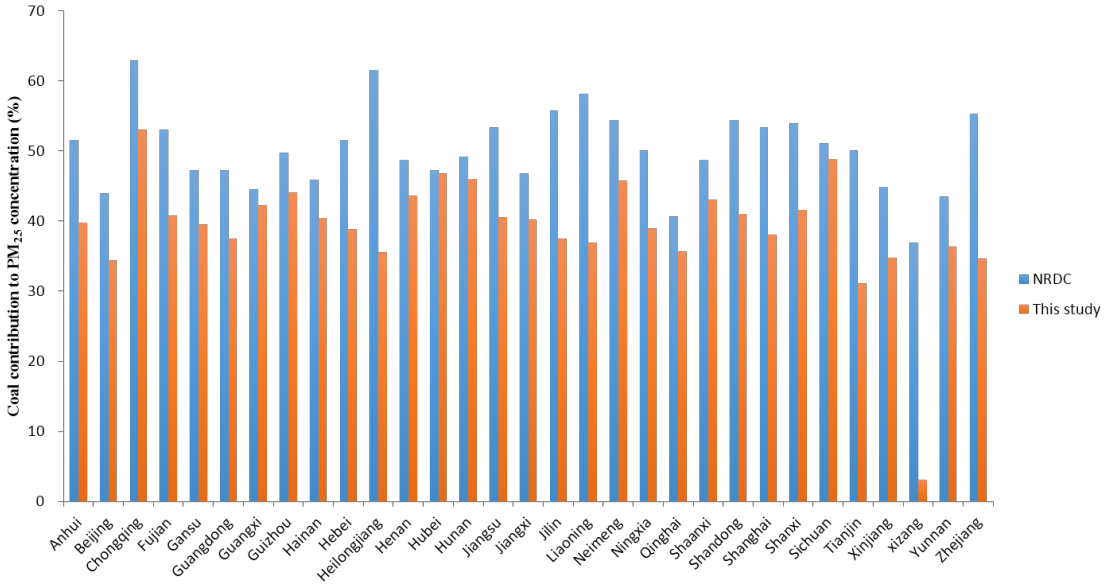


Figure 8: Seasonal contributions from coal burning in winter and summer.



~~Figure 7: Comparison of coal contribution to $PM_{2.5}$ concentration between NRDC and this study.~~

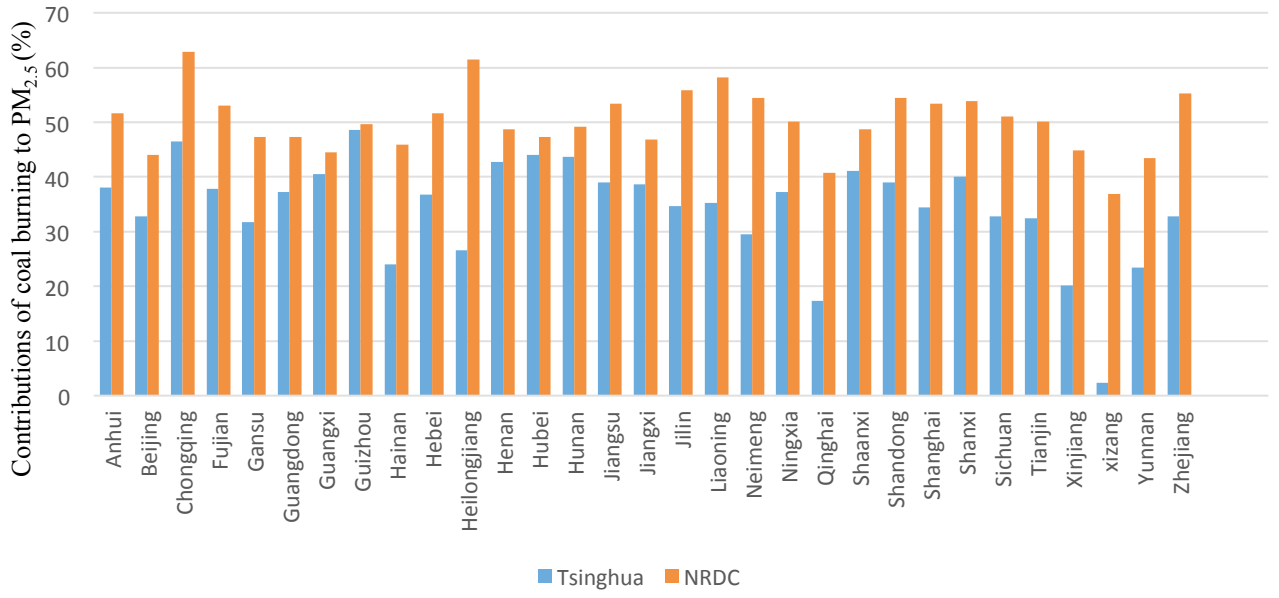


Figure 9: Comparison of coal contribution to $PM_{2.5}$ concentration between NRDC and this study.